REMOVAL OF RADIOACTIVE AEROSOLS AND GASES FROM EXHAUST AIR

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Wright-Patterson Air Force Base, Ohio

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REMOVAL OF RADIOACTIVE AEROSOLS AND GASES FROM EXHAUST AIR*

by W. Ullmann, F. Schumann, R. Schwarzbach**

SUMMARY. The National Center for Radiation Protection of the GDR (East Germany) is the national scientific center and control organ for all radiation problems coming from the peaceful use of nuclear energy, as well as questions of safety and is thus responsible for completion and expansion of knowledge on radiation protection, radiation therapy, nuclear environmental protection, nuclear-plant safety including research in the medical-biological and scientific-technological area necessary for it.

1. Introduction

The peaceful use of nuclear energy makes possible an energy supply that has a minimal impact on the environment. Present development is distinguished by a rapid increase in the number and size of plants. Thus, the danger of environmental contamination is also increased. Table I shows the radioactive exhausts of different nuclear plants currently in operation. Table II gives a survey of the predicted development to the year 2000 [1]. Compared to the natural amount of radiation (80-120 mrem/a), the given values are small. Contrary to the opinion of many authors [2] increased measures for retention of ^{85}Kr and possibly also ^{3}H may be necessary in the near future in order to counteract increased activity of these nuclei in the atmosphere. This is particularly true for densely populated countries with a high use of nuclear energy as well as for unfavorable meteorological conditions. Further development will probably lead to the determination of binding exhaust-norms for all countries.

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The program of action suggested by WHO and the IAEA [1] requires, among others, the maintenance of the safety achieved in nuclear technology, as well as the development of new and more effective methods for collection, concentration, and removal of radioactive wastes from nuclear plants, especially of ^{3}H, ^{85}Kr, and ^{129}I.

2. Survey of the Problems

Besides the mixture of radioactive products occurring from nuclear fission, activation of the cooling device or moderator can easily produce different gaseous or easily volatile compounds containing radioactive nuclei. Radioactive gases as well as radioactive aerosols and steams are present. The gaseous or steam radioactive substances or their solid reaction products remain as isolated atoms or molecules, or attach themselves to any aerosols present. Radioactive iodine, for example, can occur in aerosol form, and also in the gaseous state as elementary iodine and as organic iodine compounds.

The origination and characteristics of radioactive aerosols are, generally, far less predictable for normal operation or for disturbances; a significant portion of the radioactive exhausts is formed by radio-nuclei of gaseous or highly volatile elements. These are primarily radio-nuclei of iodine, bromine, xenon, krypton, as well as tritium. Figure 1 gives a survey of the activity and half-life of these nuclei for a nuclear power plant of 1000 MW electric output after an operating time of 2 years [3]. The calculated values for the thermal cleavage of ^{235}U are also approximately valid for rapid breeder reactors.

The amount of irradiation of the populace in the surrounding area of the nuclear plants is mostly determined by the content of the exhausts of radioactive iodine and radioactive noble gases. Contrary to this, the radioactive nuclei formed by activation are, as a rule, of subordinate importance due to their short life or their relatively small rate of formation. The problem of removal of gaseous exhausts of radioactive aerosols and gases or steams is treated below,

separately from that of radioactive aerosols, radioactive iodine, and radioactive noble gases.

3. Removal of Radioactive Aerosols

The technique of aerosol filtering has reached a high degree of development. Practically the total range of aerosol problems of nuclear plants can be solved by the previously known types of filters. In nuclear plants, high-value filters are usually used. These are the so-called suspended filters of special degree. For small requirements of degree of removal, normal suspended filters also come into consideration. Table III gives a survey of the required minimum degree of removal by suspended filters of special degree, and of suspended filters against different test aerosols frequently used [4].

TABLE I. Radioactive exhausts from nuclear plants, based on 500 MWe

Type of plant		Aerosols		Noble gas	
		MCi/a	% *)	Ci/a	% *)
Nuclear power plant	Water-pressure reactor (5 plants)	0,0037	0,6	0,02	0,7
	Boiling-water reactor (4 plants)	0,35	2	1,3	1,5
	Gas-cooled reactor (7 plants	0,03 (^{41}Ar)	12	40	—
Re-preparation plant		0,3	6,5	0,2	—

*)The percentage values are based on the ratified values.

TABLE II. Prognosis on environmental contamination by nuclear plants (according to [1]).

Prognosis for	Year	Tritium	^{129}I	^{85}Kr
Radioactive emissions	1970	< 1 MCi/a	3 Ci/a	10 MCi/a
	2000	500 MCi/a	6300 Ci/a	7000 MCi/a
Individual exposure to radiation of the population	1970	—	—	0,0004 mrem
	2000	0,04 mrem	0,2 mrem	0,4 mrem

TABLE III. Minimum degree of removal of suspended-matter filters of special degree and of suspended-matter filters (according to [4]).

Test aerosol	Oil-fog	Radioactive marked atmospheric aerosol	Quartz dust
Particle size	0,2 - 1 µm	0,01 - 0,3 µm	< 10 µm
Max. No. of particles	0,3 - 0,5 µm	0,06 - 0,12 µm	0,5 - 2 µm
Measured size	optic scattered light	Activity	optic scattered light
Suspended matter filter, special degree	> 99,70 %	(Average value > 99,90%)	Test only in special cases (always > 99%)
Suspended matter filter	> 85 %	> 70 %	Always > 95%

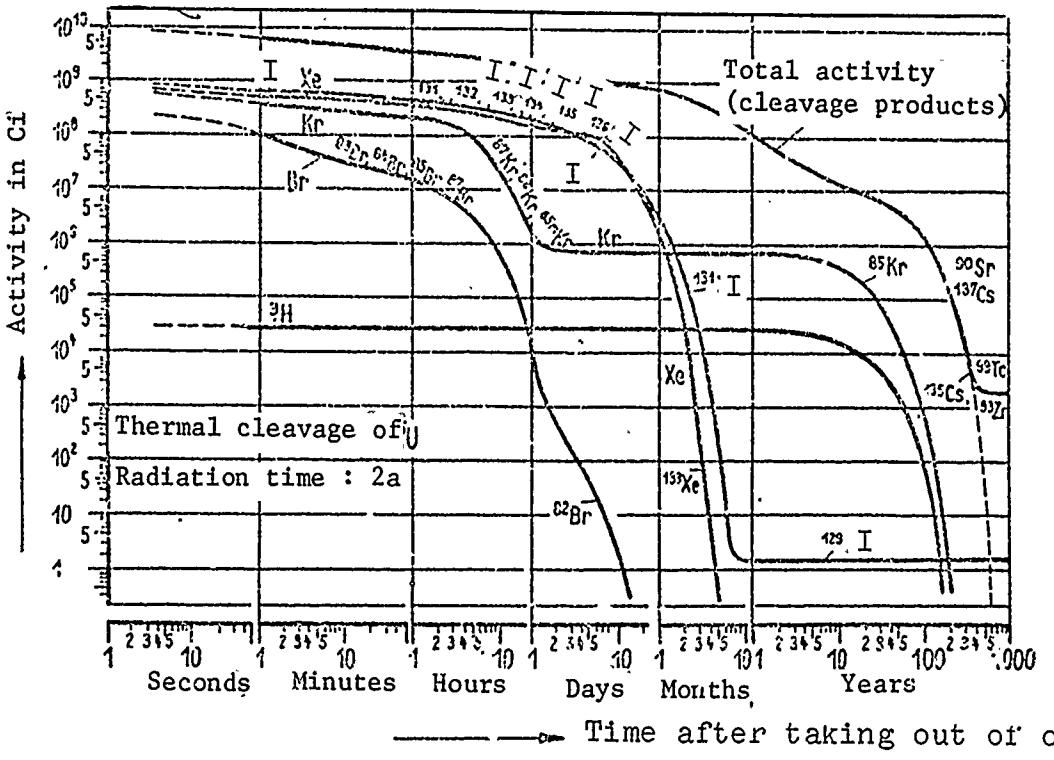

Figure 1. Decay relationships of gaseous and highly volatile radionuclei for a 1000 MWe nuclear power plant.

The following important demands are made of the aerosol filter:

a) Highest possible degree of removal at the lowest possible cost

b) Long life

The time required for the resistance to flow of the aerosol filter to double the beginning value is called the life. It is normally at least 1-2 years with sufficient pre-filtration of the air. A precondition for complete utilization of the available life is a sufficient size of the air-technological plant.

c) High operating security

The filters should be insensitive to all occurring demands. The effectiveness of filtration may not be reduced by, for example, high exhaust temperatures or by high humidity.

d) Small requirement of space

This requirement is fulfilled better by the usual square-shaped filter cells than by the so-called chamber filters which are connected to the conduit system by connection supports. However, the chamber filters have fewer problems with respect to the packing position of the filter, since their packing lines are

normally 5 to 10 times shorter than those of the filter cells.

e) The filters used must be easily removed and changed without danger

The aerosol filters currently used in nuclear plants are often the result of compromise solutions. Most nuclear plants are equipped with square-shaped filter cells. Chamber filters are used relatively seldom. In recent years, so-called well base filters [5] have been successfully used in several nuclear plants. These filters are used mostly for extreme circumstances (for example, high temperatures) and relatively small rates of flow and use mineral fibers (e.g. asbestos or basalt fibers) or mineral diffracting sands of different grandular size (e.g. quartz and basalt sands) as the filtering material. In general, suspended filters of special degree which are studded with papers containing glass fibers serve as the final step.

The optimum agreement of aerosol filters to the peculiarities of the nuclear tech ogy plant is one of the most important problems still to be solved precondition for this is, above all, the investigation of the characteristics of the radioactive aerosol to be removed. In this respect, the problem of correct evaluation of the degree of removal by aerosol filters assumes increasing importance.

If one counts on the degree of removal for nuclear technological safety criteria, then it is often not noticed that the determined degree of removal for the individual filter of the test sample does not agree with the value obtained in practice, in case the test aerosol and the aerosol present in the nuclear plant are different. The degree of removal of the total filtering apparatus normally lies significantly below the value for the individual filter, since non-seals can occur in their construction. According to previous experience, the permeability of the total plant is often 10 to 100 times higher than for the individual filters used [5].

The present conception in the GDR for the testing of filters for removal of radioactive aerosols encompasses the four steps shown in Table IV. Step I corresponds to the so-called type-testing of the individual filter. The grouping of an aerosol filter in a quantity-class means that the filter possesses the smallest capacity

established for this class. Moreover, the results obtained in the periphery of the type-test give information on the actual capacity of the filter, attained under the test conditions. They are of great importance for development of filters and for the planning of plants. For type-tested filters, the expense to the filter manufacturer (step II) for quality control, and for testing for transporting and storage damage to the operator of the filter (step III) can be kept very low. The so-called oil-fiber test is often used for testing in steps II and III [4]. With the help of this test, pores (>50μm) and tears can be determined at the damaged places by the exiting oil fibers.

The administrative test according to step IV is indispensable without use of filters with long packing lines. Table V shows the results of a total of 487 administrative tests implemented at filter-plants [6]. DOP (dioctylphthalate) was used as the test aerosol. Since the degree of removal prescribed was at least 99.95%, about 25% of the plants tested did not fulfill the requirements. The deficiency discovered by the administrative test can result from damage to the individual filters or from leaks between filter and filter clamps. If we ignore the construction of the filter in the test (step III), then an administrative test of new aerosol filters cannot determine, for example, whether the deficiencies are caused by the filter or its clamps.

The pre-conditions for an objectionless implementation of administrative tests are standard places of measurement in front of and behind the filtering unit. Complicated streaming relationships often occur at supplementally installed provisional measuring places. In these cases, the taking of a representative sample is possible only with additional expenditure of time and work.

4. Removal of Radioiodine

Because of its chemical characteristics, radioiodine can occur in different forms. Radioiodine attached to aerosols can be removed by the described aerosol filters from the exhausts of nuclear plants.

We should note here that the iodine just removed can be liberated again as gaseous iodine. The filters for gaseous radioiodine are thus situated behind the aerosol filters, generally. The pollution by aerosols of the filter medium used to separate the gaseous radioiodine is simultaneously avoided in this manner.

The filters for gaseous radioiodine were specified at first for the removal of elementary iodine. When it was demonstrated that a portion of the radioiodine was not retained by the separating device [7, 8], the existence of unknown organic iodine compounds was guessed. Organic iodine compounds were indicated in later experiments [9].

The gaseous iodine consists primarily of elementary iodine and of organic iodine compounds in the form of alkyliodides, particularly in the form of methyliodide [10]. The formation of methyliodide depends on numerous factors, among others, on the kind of gas-atmosphere and its pollutants, as well as on the temperature and the time the iodine remains in the gas. It is assumed that a portion of the methyliodide originates by reaction of elementary iodine with gaseous organic trace substances [11].

For nuclear power plants, activated charcoal filters have proven useful. Other means of absorption, e.g. silica gel or molecular sieves, are hardly used in technical standards. For removal of organic iodine compounds, the activated charcoal can be impregnated. Potassium iodide and triethylenediamine [12-14] are suitable for this. So-called reservoir filters are particularly useful. On the one hand, they make possible high sorbin layers and on the other hand, simplify the insertion of the packing in the exhaust lines because of their short packing lines. The activated charcoal filters are generally connected behind aerosol filters, in order to prevent the exit of activated charcoal particles into the exhaust system. The attainable degree of removal of 99% to 99.9% is generally sufficient for normal operation of the nuclear power plant. The iodine filters are arranged so that, even in case of damage, the ignition temperature of activated charcoal is certainly not reached. We must bear in mind the heat of disintegration of the removed radioiodine [15].

TABLE IV. Conception of the testing of filters for separation of radioactive aerosols

Step	Type of test	Implementation of testing
I	Radiation protection -- Construction test for checking the suitability of filter with respect to removal capacity, resistance to streaming, construction, and simplicity of fabrication (type test)	National control organ
II	Quality control after concluding the manufacture of the filter	Filter manufacturer
III	Testing of transport and storage damage before hook-up of the filter	Operation of the filter
IV	Testing of the total filter plant after hook-up of the filter and during its operation (administrative test).	Operation of the filter

TABLE V. Results of administrative test of 487 filtering units (according to [6])

Degree of removal of unit %	Class size		Class size		Class size 1 and 2	
	Unit with 1 or 2 filters		Unit with 3 or more filters		Total units	
	Number	%	Number	%	Number	%
100.00 – 99.97	335	76.6	15	30.0	350	71.8
99.97 – 99.95	8	1.8	4	8.0	12	2.5
99.95 – 99.90	17	3.9	4	8.0	21	4.3
99.90 – 99.0	30	6.9	20	40.0	50	10.5
99.0 – 95.0	29	6.6	4	8.0	33	6.8
95.0 – 90.0	7	1.6	1	2.0	8	1.6
90.0 – 80	4	0.9	0	0	4	0.8
80.0 – 0	7	1.6	2	4.0	9	1.8
Sum	437		50		487	

Besides activated charcoal filters, exhaust retardation plants are used. By using well-based filters, retardation times of 2-3 days, for example, can be attained [16]. Methods based on isotope exchange have mostly only been used on the laboratory scale.

On damage, one must count on the entrance of radioiodine into the rooms or in the safety enclosure of the nuclear power plant. Simultaneously, a pressure exit for the exiting steam-like primary coolant is created. The spraying plants use, for example, sodium thiosulfate, ammonium sulfate, and hydrazine [17-19]. For absorption of methyliodide at high temperatures, a hydrogen bromide solution is suitable [20].

For removal of radioiodine from the exhaust of re-preparation plants, different processes are usually combined. Besides absorption by solutions in wash-towers [e.g. with alkaline thiosulfate solution or by acidic washings using $Hg(NO_3)_2$], chemical bonding of iodine to metals (like silver) or to compounds (like silver nitrate) is utilized [14, 21]. Moreover, different types of fiber filters are used depending on the current operating conditions. In this manner, degrees of total removal of 99.99% are attained. In case the disintegration time in a future fuel-economy is reduced significantly below 100 days for prepared fuel-elements because of economic reasons, this degree of removal would no longer be sufficient. A further elevation of the degree of removal can indeed be attained by changes in technology, for example, by thorough separation of the radioiodine before the dissolving process of the fuel-elements [22]. From this proceeds the problem of developing improved processes for iodine separation from an economic point of view. This also concerns the absorption process, since, for example, the decrease of the effect at elevated gas humidities of activated charcoal is disadvantageous, as is its combustibility.

As for the testing of the iodine filters, similar requirements hold for aerosol filters. This also holds for the specifications of the positions of the required samples taken for the administrative tests, before and after iodine filters, since the molecular weight

of the organic iodine compounds is very much larger than that of the carrying gas.

5. Separation of radioactive noble gases

On normal operation of a nuclear power works, only small amounts of elemental noble gases get into the exhaust system. The much larger part remains within the fuel-element sheath and is liberated by the re-preparation of the used up fuel-elements [23]. Mostly, there are 120 to 150 days of decay time between the removal of fuel-elements from the reactor and their renewed treatment. In this time, the cleaved noble gases disintegrate, except for ^{85}Kr, into fractions of their original concentrations (see also Fig.1).

The radioactive noble gases in the exhausts of nuclear power plants consist mainly of short-lived krypton and xenon isotopes. For re-preparation plants, the long-lived ^{85}Kr is present in important amounts in the mixture with stable noble gas isotopes. Previously, it was completely expelled through the smokestacks of almost all re-preparation plants. A reduction of the decay time for the fuel elements to, say, 30 days, would not only cause significant additional difficulties for iodine removal but also for the separation of radioactive noble gases. Fig. 2 gives a survey of the most important processes for treatment and removal of radioactive cleaved noble gases from the used air of nuclear plants.

A significant reduction in the exhaust of short-lived krypton and xenon isotopes can be achieved by nuclear power plants by decay containers or retardation chambers [24]. The retardation chambers have proven valuable in practice. Economic reasons also support its use. In most cases activated charcoal is used. The absorption at room temperature can be realized by simple technical means. The requirement of space is, however, significantly greater than for low-temperature absorption (cooling with liquid nitrogen). The activated charcoal filters are normally so constructed that the radionuclei of xenon are held back up to 30 days. The retardation time for krypton is significantly lower, however, it is sufficient for the decay of the short-lived krypton isotopes.

Adsorption processes are also used for re-preparation plants, for example, for fine purification and final concentration in combination with a liquid-absorption process [23]. The process, developed in recent years, of liquid-absorption using freon 12 is already technically ready for use [25]. For the future, it looks very promising. The use for pre-treatment of exhausts is relatively small. Krypton, too, is separated very well.

The process of membrane diffusion has only been tested in the laboratory. This process uses the good solubility of noble gases in high polymers [26]. By using thin membranes of methyl phenyl silicon rubber, it is possible to separate krypton and xenon from other gases. The process could be expanded to purify the protective gas of rapid breeder reactors as well as for purification of exhaust in re-preparation plants. By using the results of experimental investigations on membranes, calculations for the use of this process have already been implemented in the technical arena [27].

Cleaved noble gases can also be separated from liquified exhausts by fractional distillation. The process offers relatively small investment and operating costs. A disadvantage is the danger of explosion as the consequence of ozone formation. The processes of thermal and electrostatic diffusion are not technically suitable for the removal of radioactive noble gases.

In the future, fixation and safe disposal of cleaved noble gases will increase in importance. Enclosing radioactive noble gases in solid bodies in the form of inclusive compounds (clathratation) or valence compounds appears very advantageous; however, it still requires procedural development [28, 29]. The disposal can be done, for example, in special containers above or below ground. Also, disposal of radioactive gases or of highly contaminated used-air streams in geological formations [30] is of great importance to the nuclear industry, as well as for safety and economic reasons. This process appears particularly promising as a solution to the industrial problem of reappearing exhausts from re-preparation plants.

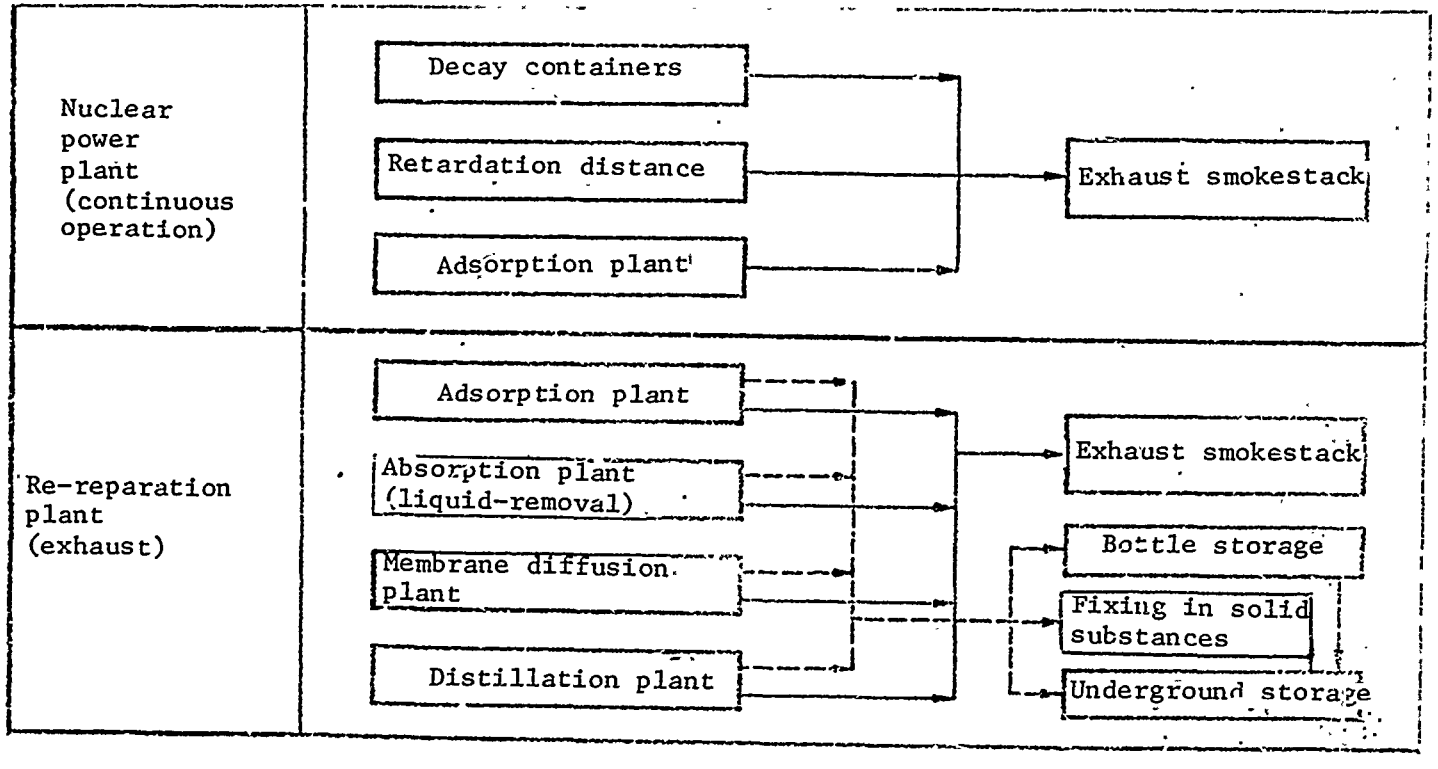

Figure 2. Treatment and Disposal of radioactive cleaved noble gases (schematic)

Further development in the area of separation of ^{85}Kr depends very much on progress in the work of establishing internationally binding exhaust norms.

6. Conclusions

The present technical state attained in the area of purification of gaseous exhausts from nuclear plants makes it possible to protect the environment of nuclear plants sufficiently from radioactive aerosols, gases, and steams. This is one of the important advantages which nuclear plants have over conventional plants with respect to environmental influence due to gaseous exhausts. For the future, a favorable prognosis can be given, too. On the one hand, the purification processes for radioactive exhausts are still sufficiently susceptible to development; on the other hand, possibilities are indicated for an advantageous operation of, for example, re-preparation plants. In connection with modern methods of prolonged storage, future problems may also be solved in an economical and safe manner.

An important proposition is the further development and international agreement on different measuring and control methods. This concerns primarily the technical supervision of radioactive exhausts as well as testing of filters including administrative examination of the purification plants. The education and use of experts for the problems of air technology and air purification encountered in nuclear plants should be given greater attention now.

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